First Separate Measurements of the Nondipole Parameters γ and δ: Showcase Neon 2p Photoemission

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INTRODUCTION

The familiar dipole approximation [i.e., exp(ik·r)≈1] limits photon interactions to purely electric-dipole (*E1*) effects. For photoemission processes, this approximation leads to the well-known expression for the differential photoionization cross section,

$$\frac{d\sigma(h\nu)}{d\Omega}=\frac{\sigma(h\nu)}{4\pi}\left[1+\frac{\beta(h\nu)}{2}\left(3\cos^2\theta-1\right)\right] \qquad (1)$$

which describes the angular distribution of photoelectrons ejected from a randomly oriented sample by 100 % linearly polarized light. Here, $\sigma(h\nu)$ is the partial photoionization cross section, and θ is defined as the angle between the outgoing electron and the photon polarization vector of the incoming, linear polarized light. In the dipole approximation, the parameter $\beta(h\nu)$ completely describes the angular distribution of photoelectrons, and all higher-order interactions, such as electric-quadrupole (*E2*) and magnetic-dipole (*M1*), are neglected. Over the past few decades, the dipole approximation has facilitated a basic understanding of the photoionization process in atoms and molecules, as well as the application of photoelectron spectroscopy to a wide variety of condensed-phase systems.

Beyond the dipole approximation, predictions and observations of high-photon-energy ($h\nu \geq 5$ keV) deviations from dipolar photoelectron angular distributions have enjoyed a successful history dating from the 1930's. Small deviations from expected dipolar angular distributions at photon energies between 1 and 2 keV were attributed qualitatively to the effects of higher-order photon interactions. These so-called *nondipole* effects in the angular distributions of photoelectrons, which are primarily due to first-order (*E2* and *M1*) corrections [*O(k)*] to the dipole approximation [i.e., exp(ik·r) ≈1 + ik·r], were later shown to lead to the expression

$$\frac{d\sigma(h\nu)}{d\Omega}=\frac{\sigma(h\nu)}{4\pi}\left[1+\frac{\beta(h\nu)}{2}\left(3\cos^2\theta-1\right)+\left(\delta(h\nu)+\gamma(h\nu)\cos^2\theta\right)\sin\theta\cos\phi\right] \qquad (2)$$

for 100 % linearly polarized light. The angle θ is defined above and ϕ is the angle between the photon propagation vector and the projection of the electron momentum vector into the plane perpendicular to the photon polarization vector. $\gamma(h\nu)$ and $\delta(h\nu)$ are the nondipole angular-distribution parameters attributable to first-order interactions only. Values of the first-order nondipole parameters are determined by the strength of electric-quadrupole (E2) and magnetic-dipole (M1) photoionization amplitudes relative to the corresponding electric-dipole (E1) amplitudes. Expressions for γ and δ include *only* cross terms of the E2 and M1 amplitudes with the E1 amplitudes; "pure" quadrupole or magnetic-dipole interactions are not present in the first-order correction. Because the pure transitions are not included, the only manifestation of the first-order breakdown in the dipole approximation will be a change in the angular distribution of photoelectrons, specifically a forward/backward asymmetry relative to the photon propagation direction. The initial experiments motivated theoretical work, and more recent publications include quantitative predictions for a variety of atomic subshells. To date, all published experimental nondipole data did not separate the γ and δ parameters, except for the special case of n*s* photolines,

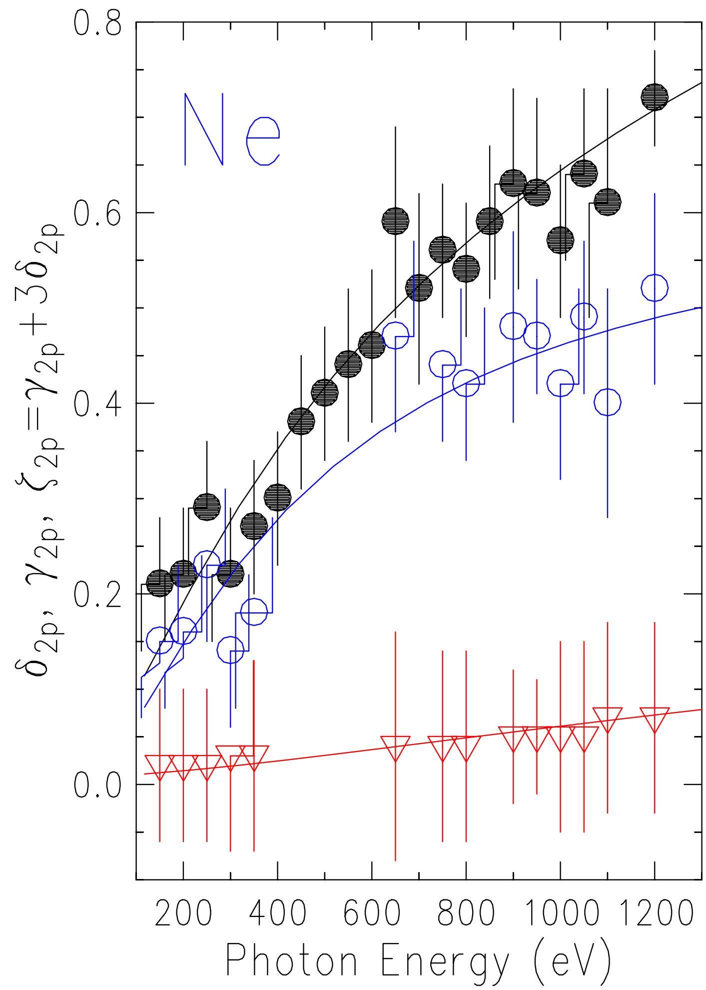

Figure 1. Photoelectron angular-distribution anisotropy parameter, δ (open triangles), γ (open circles), and ζ (filled circles), for Ne 2*p*. The theoretical data (solid line) are independent-particle calculations.

where δ=0. The sum of γ and δ parameters is commonly expressed using the ζ parameter (ζ = γ + 3δ), which has been used for most of the experimental and recent theoretical work. The present work concentrates on the separate determination of γ and δ parameters for Ne 2*s* and 2*p* valence photoemission between 150 and 1200 eV and comparison to theoretical data.

EXPERIMENTS

The present experiments were performed at the Advanced Light Source (ALS) of the Lawrence Berkeley National Laboratory on undulator beamline 8.0, which covers the 80-1300 eV photon-energy range. During the measurements the ALS operated at 1.9 GeV in two-bunch mode with a photon pulse every 328 ns. Four time-of-flight (TOF) electron analyzers, equipped with microchannel-plate detectors, collected spectra simultaneously at different angles. The interaction region was formed by an effusive gas jet intersecting the 0.4-mm-diameter photon beam. Energy resolution of the TOF analyzers with a focus size of 0.4 mm is about 1% of the final electron kinetic energy. A retarding voltage was used to slow the high-kinetic-energy electrons to a final kinetic energy of about 200 eV, thus maintaining the analyzer resolution at about 2 eV.

RESULTS

Figure 1 shows the experimental and theoretical data for the γ, δ, and ζ parameters for Ne 2*p* photoionization. All data agrees very well with theory but for certain energies it was not possible to satisfactorily separate the two parameters γ and δ and therefore the separated nondipole components γ and δ show less data than there are shown for ζ. Even though δ is very small for the Ne 2*p* photoionization it is not zero and clearly shows a trend to increase with photon energy. The relatively large error bars reflect a possible systematic uncertainty and not the statistical error of each point.

CONCLUSIONS

In summary, this is the first comprehensive study of dipole and nondipole angular-distribution parameters for atomic valence photoionization, in this case covering the photon-energy region from 150 eV to 1200 eV. For the first time the individual nondipole parameters γ and δ have been separately determined for a non-*s*-subshell photoline. The measured nondipole contributions to the photoelectron angular distributions agree very well with existing calculations. It is important to appreciate the fact that doing gas-phase and solid-state angle-resolved photoemission experiments can show sizeable nondipole effects below 1 keV; this work clearly demonstrates nondipole effects may need to be considered in photoemission measurements, *even* for $h\nu<1$ keV.

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